

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re application of:

Christoph Briehn et al.

Serial No.: 10/599,285

Filed: September 25, 2006

For: CURABLE COMPOSITION CONTAINING  
SURFACE-MODIFIED PARTICLES

Attorney Docket No.: WAS 0807 PUSA

Group Art Unit: 4171

Examiner: Hannah J. Pak

**APPEAL BRIEF UNDER 37 C.F.R. § 41.37**

Mail Stop Appeal Brief - Patents  
Commissioner for Patents  
U.S. Patent & Trademark Office  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

This is an Appeal Brief from the final rejection of claims 11 - 18 of the Office Action mailed on February 12, 2009 for the above-identified patent application.

**I. REAL PARTY IN INTEREST**

The real party in interest is Wacker Chemie AG ("Assignee"), a corporation organized and existing under the laws of Germany, and having a place of business at Hanns-Seidel-Platz 4, Muenchen, Germany D-81737, as set forth in the assignment recorded in the U.S. Patent and Trademark Office on May 21, 2007 at Reel 019728/Frame 0028.

**II. RELATED APPEALS AND INTERFERENCES**

There are no appeals, interferences or judicial proceedings known to the Appellant, the Appellant's legal representative, or the Assignee which may be related to, directly affect or be directly affected by or have a bearing on the Board's decision in the pending appeal.

### III. STATUS OF CLAIMS

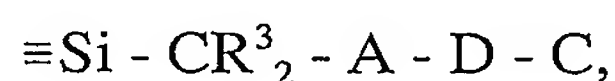
Claims 11 - 18 are pending in this application. Claims 11 - 18 have been rejected and are the subject of this appeal. Claims 1 - 10 have been cancelled.

### IV. STATUS OF AMENDMENTS

An amendment after final rejection was filed on June 4, 2009, and has been accepted for entry. No claim amendments have been made.

### V. SUMMARY OF CLAIMED SUBJECT MATTER

The invention of claim 11 is directed to curable compositions containing a binder bearing at least one ethylenically unsaturated group, and functionalized particles bearing at least one ethylenically unsaturated group (page 1, lines 3 - 7; page 4, lines 21 - 25), which contains radicals of the formula



prepared by reacting particles containing reactive Me-OH, Si-OH, Me-O-Me, Me-O-Si, Si-O-Si, Me-OR<sup>1</sup> or Si-OR<sup>1</sup> groups (page 5, lines 22 - 24) with organosilanes of the formula



their hydrolysis or condensation products or mixtures thereof (page 5, lines 25 - 29) and optionally water, where R<sup>1</sup> is hydrogen or a C<sub>1-6</sub> hydrocarbon radical whose carbon chain is optionally interrupted by non-adjacent oxygen, sulfur, or NR<sup>4</sup> groups (page 5, line 32 to page 6, line 1); R<sup>2</sup> is a C<sub>1-12</sub> hydrocarbon radical optionally interrupted by non-adjacent oxygen, sulfur or NR<sup>4</sup> groups (page 6, lines 2- 4); R<sup>3</sup> has the same definition as R<sup>2</sup> (page 4, lines 31 - 33); R<sup>4</sup> is hydrogen or a C<sub>1-12</sub> hydrocarbon radical (page 5, lines 1- 2); A is oxygen, sulfur, =NR<sup>4</sup>, or = N-(D-C) (page 5, line 3); D is a carbonyl group, alkylene, cycloalkylene, or arylene radical

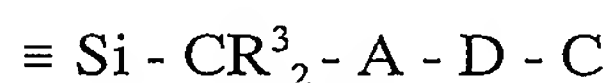
having 1 - 12 carbon atoms optionally interrupted by non-adjacent O, S, or =NR<sup>4</sup> groups (page 5, lines 4 - 8); C is an ethylenically unsaturated group (page 5, line 9); Me is a metal atom (page 6, line 5); and n is 0, 1, or 2 (page 6, line 6).

## **VI. GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL**

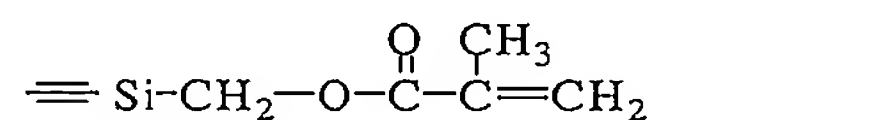
Claims 11 - 18 stand rejected under 35 U.S.C. § 103(a) over Olsen et al. U.S. Patent No. 4,491,508 (hereinafter, "*Olsen*").

## **VII. ARGUMENT**

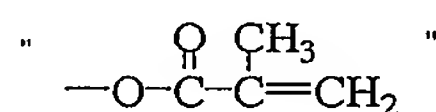
The present invention pertains to curable compositions containing a binder with ethylenic unsaturation and a unique filler ("functionalized particles") which also bears ethylenic unsaturation. The functionalized filler particles offer highly improved scratch resistance, and are easy and economical to prepare. The fillers are prepared by reacting a silica or silicon/mixed metal (Me) oxide with a functionalizing reagent



where C is an unsaturated group (the remaining R<sup>3</sup>, A, and D moieties have been delineated earlier). The most preferred functionalizing compounds have R<sup>1</sup> = methyl, R<sup>3</sup> = hydrogen, A = oxygen, and D = carbonyl, and thus have the structure:



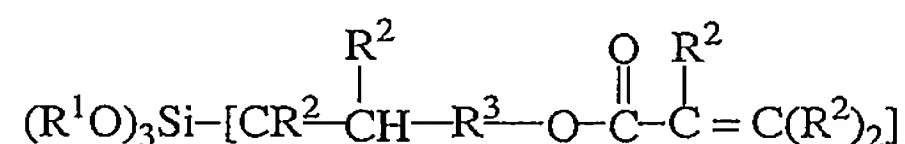
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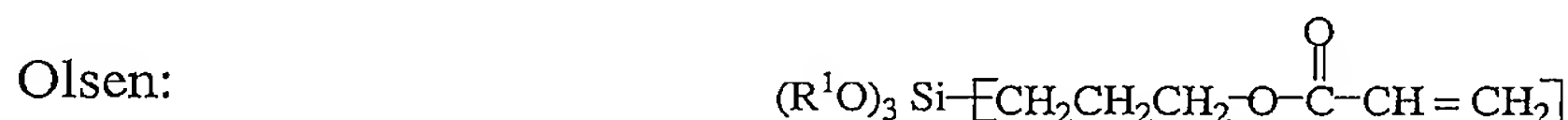
radical is a methacrylato radical. Note, most importantly, that the silicon is separated from the methacrylato group by a single carbon atom, in the formula above, a methylene group<sup>1</sup>. These compounds are thus " $\alpha$ -silanes" where the functional group is  $\alpha$  to the silicon.

The Office contends that the compounds used by *Olsen* are structurally similar to those used as claimed by Applicants. However, this is incorrect.

*Olsen's* alkoxysilanes, have the formula (simplified) from column 1 and claim 1:



The Office states that this compound, when  $R^2 = H$ , and  $R^3 = C_{1-8}$  hydrocarbon radical, is similar to Applicants' compounds when Applicants  $R^3 = a C_{1-12}$  hydrocarbon radical. This is incorrect, however. When these substitutions are made, and  $R^3$  in *Olsen* is a methylene group, and  $R^3$  of Applicants is a methyl group, the structures are as follows:



Note that while in *Olsen*, the "spacer" group between the silicon and oxygen is a propylene group (minimally), the spacer in Applicants' compound is still a methylene group, in this case a dimethylmethylene group. If  $R^3$  were a  $C_6$  hexyl group, in *Olsen*, the spacer would an octylene

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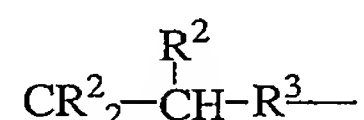
<sup>1</sup>In less preferred embodiments, by a substituted methylene group. In every case, however, silicon is separated from the functional group containing the ethylenic unsaturation by but a single carbon atom, i.e. a "methylene" carbon atom.

group, where as in Applicants' compound it would be a dihexylmethylene group, *i.e.* there is still only one carbon between Si and O.

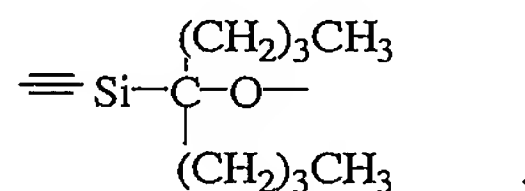
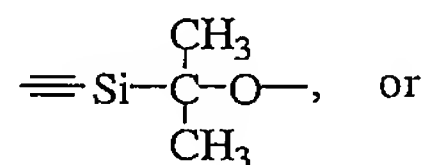
When in Applicants' formula  $R^3 = H$ , Applicants' compound is:



As can be clearly seen, the



group of *Olsen* requires minimally a chain of three carbon atoms, since  $R^3$  is a  $C_{1-8}$  alkylene group ( $-CH_2-$ ,  $-CH_2CH_2-$ ,  $-CH_2CH_2CH_2-$ , etc.) (*Olsen* column 1, line 66: " $R^3$  is a divalent  $C_{1-8}$  alkylene radical"). Thus, in *Olsen*'s compounds, the silicon of the alkoxysilyl group is separated from the O of the methacrylate group by minimally three carbon atoms, *e.g.* 2-methacrylatopropyltrimethoxysilane, used in all of *Olsen*'s examples. In Applicants' composition, the silicon of the alkoxysilyl group is separated from the methacryloxy group by but a single carbon atom, regardless of whether this carbon atom is substituted by hydrogen or an alkyl group, *i.e.*



As indicated in MPEP § 2144.09, a rejection for obviousness is only appropriate in cases of close structural similarity when the respective compounds are position isomers (*i.e.* 1,3-dichlorophenyl versus 1,2-dichlorophenyl) or adjacent homologs, *i.e.* differing by but a single  $-\text{CH}_2-$  group. Moreover, *prima facie* obviousness based on structural similarity is rebutted when unexpected results are obtained.

With respect to homology, it is well known in Chemistry that there may be great differences in compounds having methylene "spacer" groups as compared with otherwise similar compounds having ethylene, propylene, or butylene "spacers." This is not a case such as in the pharmaceutical arts where differences between a compound substituted with an ethyl group is only qualitatively different from one substituted with a methyl or propyl group. Here, the methylene "spacer" group separates a reactive functional group from a semi-metal silicon atom. In such cases, differences in inductive effects and anchimeric assistance come into play, as is well known. See, *e.g.* March, *infra*, a well known Organic Chemistry treatise.

Moreover, case law dictates that homology itself is not controlling, and must be considered in view of the knowledge of the art as a whole. See, *e.g.* *In re Mills*, 281 F.2d 218, 126 USPQ 513 (CCPA 1960) and *In re Wiechert*, 370 F.2d 927, 152 USPQ 247 (CCPA 1967), which indicate that "all other relevant facts must be considered."

Here, the minimum size of *Olsen's* "linking" group between Si and O is propyl,  $-\text{CH}_2\text{CH}_2\text{CH}_2-$ , and the maximum is decyl  $-(\text{CH}_2)_{10}-$ . In Applicants' process, the maximum length linking group is methylene,  $-\text{CH}_2-$ . Methylene and propylene are not adjacent homologs, and the principles of homology and structural similarity do not apply. Moreover, in Applicant's compounds, anchimeric assistance or inductive effects may occur, while neither anchimeric assistance nor inductive effects can operate with propylene, or higher alkylene linking groups. See, *e.g.*, March, ADVANCED ORGANIC CHEMISTRY: REACTIONS, MECHANISMS AND STRUCTURE, McGraw-Hill, New York, c. 1968, p. 362.

Moreover, compounds containing an alkoxysilyl group separated from an electronegative group by but a methylene group are known to behave very differently from otherwise similar compounds containing a propylene group. For example, aminopropyltrimethoxysilane is a compound which can be dispersed in water and hydrolyzes by reaction of the silicon-bonded methoxy groups to produce polymeric and oligomeric siloxanes, whereas aminomethyltrimethoxysilane, as reported in the scientific literature, decomposes in water, liberating methyl amine and producing silica, SiO<sub>2</sub>. These compounds are far different, the difference being the nature of the linking group: methylene or propylene.

In Applicants' last response and as reiterated above, one skilled in the art is thus aware that  $\alpha$ -silanes, *i.e.* silanes having but a single methylene "spacer" between the semi-metal silicon atom and a functional group react far differently from otherwise similar compounds with, *e.g.* propyl spacers. Reference may be had to Adima et al., "Facile Cleavage of Si-C Bonds During the Sol-Gel Hydrolysis of Aminoethyltrialkoxysilanes - A New Method for the Methylation of Primary Amines," EUR. J. ORG. CHEM., 2004, 2582 - 2588, which indicates, as recited previously, that while propyl-spaced aminoalkyltrialkoxysilanes react in sol-gel hydrolysis to provide aminopropyl-functional silsesquioxanes, attempts to prepare "homologous" aminomethyl-functional compounds failed. Rather, scission at the Si-C bond occurred, producing silica and the corresponding substituted amine. This is conclusive evidence that  $\alpha$ -silanes are in fact very different from  $\beta$ - or  $\gamma$ -silanes, and the principle of homology fails for these compounds.

Dr. Christoph Briehn, one of the inventors of the claimed invention, prepared additional examples and comparative examples. In these examples, the proportion of silica used is kept at 35 weight percent. In the first example (inventive), the claimed methacrylatomethyltrimethoxysilane<sup>2</sup> is used to functionalize the silica, whereas in Comparative Examples A and B, an *Olsen* silane, methacrylatopropyltrimethoxysilane was used. In

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<sup>2</sup>With a methylene spacer between Si and O



Comparative Example A, the solvent used was t-butanol, as used by *Olsen*. In Comparative Example B, ethanol, as used in the additional inventive example, was employed. The scratch resistance of the comparative example using the same solvent (ethanol) was 26% lower than the inventive example, while that of the comparative example using *Olsen*'s t-butanol solvent was lower by 21%. A 20 plus % increase in scratch resistance is highly significant, and both surprising and unexpected. *Olsen* does not teach or suggest using Applicants' claimed silanes to functionalize silica, and the results achieved are highly surprising and unexpected. Withdrawal of the rejection of the claims over *Olsen* for both these reasons is respectfully solicited.

The Declaration of Dr. Briehn also indicates (¶¶s 2, 3, page 1), and (¶5, pages 3 - 4), that there has been a long felt need in the industry to provide increase scratch resistance, which anyone having taken a new car through several contact-type car washes knows, since numerous fine scratches on the freshly painted vehicle occur. Dr. Briehn indicates that the subject invention solved the long felt need for higher scratch resistance by the claimed composition, despite the fact that the *Olsen* reference issued in 1985, more than 20 years ago. Despite this long felt need, no one has disclosed, taught, or suggested the claimed composition and its unique functionalized fillers. These secondary considerations, long felt need and failure of others, must be considered. The Office has not disputed this evidence provided by Dr. Briehn, one having considerable skill in the art.

Moreover, the examples and comparative examples bear this out. The inventive examples had a loss of gloss in the Peter-Dahn scratch test which averaged <17.7 whereas the comparative examples averaged 42, an improvement of almost 200% (a factor of almost 3). These improved results are clearly surprising and unexpected and clearly rebut any contention of *prima facie* obviousness.



Reversal of the rejections of record is earnestly and respectfully solicited.

The fee of \$540.00 as applicable under the provisions of 37 C.F.R. § 41.20(b)(2) is being charged to Deposit Account No. 02-3978 via electronic authorization submitted concurrently herewith. The Commissioner is hereby authorized to charge any fees or credit any overpayments as a result of the filing of this paper to Deposit Account No. 02-3978.

Respectfully submitted,

**Christoph Briehn et al.**

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Enclosure - Appendices

### VIII. CLAIMS APPENDIX

Claims 1 - 10. (Cancelled)

11. A curable composition comprising a binder bearing at least one ethylenically unsaturated group, and functionalized particles which possess at least one ethylenically unsaturated group and contain radicals of the formula I,



where the particles are prepared by reacting

(a) particles of a metal oxide, metal-silicon mixed oxide, silicon dioxide, colloidal silicon dioxide, organopolysiloxane resin or combination thereof, the particles possessing functionality selected from Me-OH, Si-OH, Me-O-Me, Me-O-Si-, Si-O-Si, Me-OR<sup>1</sup> and Si-OR<sup>1</sup>, and having an average diameter of less than 1000 nm,

(b) with organosilanes of the general formula II,



their hydrolysis or condensation products, or mixtures thereof, and

where

(c) and optionally water,

- $R^1$  is hydrogen or a hydrocarbon radical having 1 to 6 carbon atoms, whose carbon chain is optionally interrupted by nonadjacent oxygen, sulfur or  $NR^4$  groups,
- $R^2$  is a hydrocarbon radical having 1 to 12 carbon atoms, whose carbon chain is optionally interrupted by nonadjacent oxygen, sulfur or  $NR^4$  groups,
- $R^3$  is hydrogen or a hydrocarbon radical having 1 to 12 carbon atoms, whose carbon chain is optionally interrupted by nonadjacent oxygen, sulfur or  $NR^4$  groups,
- $R^4$  is hydrogen or a hydrocarbon radical having 1 to 12 carbon atoms,
- A is oxygen, sulfur,  $=NR^4$  or  $=N-(D-C)$ ,
- D is a carbonyl group, or an alkylene, cycloalkylene or arylene radical having 1 to 12 carbon atoms, the carbon chain optionally interrupted by nonadjacent oxygen, sulfur or  $NR^4$  groups,
- C is an ethylenically unsaturated group,
- Me is a metal atom, and
- n is 0, 1 or 2.

12. The composition of claim 11, wherein the particles are selected from pyrogenic silica, colloidal silica, and silicone resins.

13. The composition of claim 11, wherein the hydrocarbon radical  $R^1$  is a methyl, ethyl or phenyl radical.

14. The composition of claim 11, wherein at least one group (-A-D-C) is a radical  $\text{OC(O)C(CH}_3\text{)=CR}_2^3$ ,  $\text{OC(O)CH=CR}_2^3$ ,  $\text{NHC(O)C(CH}_3\text{)=CR}_2^3$ , or  $\text{NHC(O)CH=CR}_2^3$ .

15. The composition of claim 11, wherein the ethylenically unsaturated groups in the binder are capable of free-radical, cationic or anionic polymerization.

16. The composition of claim 11, wherein the ethylenically unsaturated groups in the binder are polymerizable by actinic radiation or thermal treatment.

17. The composition of claim 11, wherein the ethylenically unsaturated groups in the binder are selected from vinyl groups, methacrylate groups, acrylate groups, acrylamide groups, and mixtures thereof.

18. The process for coating a substrate, comprising applying to a surface of said substrate a composition of claim 11, and curing said composition.

**IX. EVIDENCE APPENDIX**

1. Declaration of Dr. Briehn.

**X. RELATED PROCEEDINGS APPENDIX**

None.